THE PREDICTION OF OSMOTIC AND ACTIVITY COEFFICIENTS FOR ELECTROLYTE MIXTURES AT ELEVATED TEMPERATURES

M. H. Lietzke and R. W. Stoughton

MASTER

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Printed in the United States of America. Available from
National Technical Information Service
U.S. Department of Commerce
5285 Port Royal Road, Springfield, Virginia 22151
Price: Printed Copy \$4.00; Microfiche \$1.45

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Contract No. W-7405-eng-26

CHEMISTRY DIVISION

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SEPTEMBER 1974

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ABSTRACT

In fields such as geothermal energy and desalination it is necessary to have thermodynamic information on aqueous mixed electrolyte solutions over wide ranges of temperature. Direct measurements, particularly above 100°, are difficult experimentally and very time consuming. Hence, it is desirable to develop methods for predicting properties such as osmotic and activity coefficients for electrolyte mixtures at elevated temperatures. In this report we demonstrate that simple expressions developed for predicting such quantities for electrolyte mixtures at 25° function equally well at elevated temperatures. Because of the success of these predictions we feel that the techniques used in the calculations presented in this report could be used to predict closely osmotic and activity coefficient behavior in as yet unstudied systems, such as the solutions of geothermal interest.

INTRODUCTION

Fields such as geochemistry, desalination, and oceanography have been receiving increased attention in recent years. In each of these areas one encounters the need for information on the thermodynamic properties of aqueous electrolyte mixtures. One of the problems that has attracted considerable attention is the prediction of the activity coefficient of each component (electrolyte) in an aqueous solution containing several electrolytes.¹ Another is the prediction of the osmotic coefficient of an aqueous solution containing several electrolytes.² Some of these prediction techniques are very elaborate and require the use of a high speed computer, while others involve the use of very simple equations. In all cases, however, predictions of osmotic and activity coefficients for electrolyte mixtures have been reported at 25° only.

¹G. Scatchard, R. M. Rush, and J. S. Johnson, *J. Phys. Chem.*, 74, 3786 (1970); P. J. Reilly, R. H. Wood, and R. A. Robinson, *ibid.*, 75, 1305 (1971); M. H. Lietzke and R. W. Stoughton, *J. Solution Chem.*, 1, 299 (1972); Kenneth S. Pitzer and Janice J. Kim, LBL-2713, (March 1974).

²M. H. Lietzke and R. W. Stoughton, *J. Inorg. Nucl. Chem.*, 36, 1315 (1974).

It is now becoming apparent, in fields such as desalination and geothermal energy, that thermodynamic information is needed on electrolyte mixtures at elevated temperatures. Direct measurements, particularly above 100°, are difficult experimentally and very time consuming. Hence, it seems desirable to develop methods for predicting properties such as osmotic and activity coefficients for electrolyte mixtures at elevated temperatures. Such values need not be of the highest possible accuracy; values good to 1-2% are entirely adequate for practical applications.

In developing mathematical models for predicting thermodynamic properties of mixed systems it is desirable to have some experimental data on the systems against which to check the predictions. Unfortunately, the number of aqueous electrolyte mixtures that has been studied experimentally at elevated temperatures is very small. By means of the emf technique we have investigated a series of solutions containing HCl and various metal chlorides to temperatures from 75 to over 200°. ³ In addition, the osmotic coefficient of synthetic sea water has been measured to 100° in one study ⁴ and to 300° in another ⁵. As far as simple electrolyte solutions are concerned only the osmotic coefficients of HCl ⁶ NaCl, ^{5,7,8} MgCl₂, ⁵ and MgSO₄ ⁵ have been carefully and extensively measured to elevated temperatures.

³M. H. Lietzke and R. W. Stoughton, J. Phys. Chem. 69, 2395 (1965); 70, 756 (1966); 71, 662 (1967); 72, 257 (1968); 72, 4408 (1968); J. Tenn. Acad. Sci. 44, 66 (1969); J. Inorg. Nucl. Chem. 31, 3481 (1969); 33, 1649 (1971); 34, 2233 (1972); 34, 3789 (1972); 35, 1651 (1973); J. Chem. Eng. Data 17, 459 (1972).

⁴H. Frank Gibbard, Jr., and George Scatchard, J. Chem. Eng. Data 17, 498 (1972).

⁵Chia-tsun Liu and W. T. Lindsay, Jr., Int-OSW-RDPR-71-722, U. S. Dept. of the Interior, December 1971.

⁶R. S. Greeley, W. T. Smith, Jr., M. H. Lietzke, and R. W. Stoughton, J. Phys. Chem. 64, 1445 (1960).

⁷E. R. Gardner, P. J. Jones, and H. J. DeNordwall, Trans. Faraday Soc. 59, 1994 (1963).

⁸George Scatchard, H. Frank Gibbard, and Raymond A. Rousseau, unpublished work.

Reference has previously been made ^{1,2} to papers in which we demonstrated that relatively simple expressions could be used to predict osmotic and activity coefficient values for electrolyte mixtures at 25°C. In this report we will show that these equations function equally well at elevated temperatures. Because of the paucity of mixture data at elevated temperatures we will restrict ourselves to the prediction of the osmotic coefficient of synthetic sea water to 300° and of the activity coefficient of each component in HCl-NaCl-MgCl₂ mixtures to 55°. Because of the success of these predictions we feel that the techniques used in these calculations could be used to predict similar quantities for as yet unstudied systems, such as the solutions of geothermal interest.

METHOD OF CALCULATION

In carrying out the calculations osmotic coefficient data for HCl, NaCl, MgCl₂, and MgSO₄ solutions at 25°⁹ were combined with data at higher temperatures from references (5), (6), (7), and (8). These sets of osmotic coefficient data were then fitted by the method of least squares using

equation (1):

$$\phi = 1 - \frac{|z_+ z_-| S_T}{(1.5)^3 I} \left[1 + 1.5\sqrt{I} - 2 \ln (1 + 1.5\sqrt{I}) - \frac{1}{1 + 1.5\sqrt{I}} \right] + BI + CI^2 \quad (1)$$

In this equation ϕ is the osmotic coefficient; z_+ and z_- represent the charges on the cation and anion respectively; S_T is the Debye-Hückel limiting slope at absolute temperature T ; and I is the ionic strength of the solution on a molal basis. The coefficients B and C are temperature dependent and were assumed to vary with temperature as shown in equations (2) and (2a). The form of these latter equations is consistent with the

$$B = p_1 + p_2/T + p_3 \ln T \quad (2)$$

$$C = p_4 + p_5/T + p_6 \ln T \quad (2a)$$

assumption that $\bar{C}_p$ for each electrolyte is constant over the temperature range of the data.

⁹R. A. Robinson and R. H. Stokes, Electrolyte Solutions, Academic Press, Inc., New York, 1955.

When equations (2) and (2a) expressing the temperature dependence of the coefficients $\underline{B}$ and $\underline{C}$ were inserted into equation (1) and the least squares fits performed, the values of the parameters $\underline{p}_1$ shown in Table 1 were obtained. The temperature range of the osmotic coefficient data used in the least squares fits was 25 - 300° in the case of NaCl and MgCl₂ and 25 - 200° in the case of HCl and MgSO₄. With the parameters listed in Table 1 equation (1) can be solved to give the value of the osmotic coefficient of a solution of any of the four electrolytes at any particular combination of temperature and ionic strength.

THE OSMOTIC COEFFICIENT OF SYNTHETIC SEA WATER FROM 25° TO 300°

In a previous paper¹ we demonstrated that the equation

$$\phi_m = \frac{\sum_{j=2}^n v_j m_j \phi_j}{\sum_{j=2}^n v_j m_j} \quad (3)$$

could be used to make remarkably good predictions of the osmotic coefficient of a variety of aqueous electrolyte mixtures. In this equation ϕ_m represents the osmotic coefficient of an aqueous solution containing n electrolytes each indexed as $j = 2, 3, \dots, n$, the subscript 1 being reserved for the solvent. v_j is the number of ions into which the j th electrolyte dissociates, m_j is the corresponding molality of the electrolyte in the mixture, while ϕ_j represents the osmotic coefficient of a solution of the pure electrolyte at the total ionic strength of the solution. When equation (3) is used to predict at 25° values of the osmotic coefficient of synthetic sea water containing NaCl-MgCl₂-MgSO₄ in the ratio 1 : 0.0745 : 0.0586 the results shown in Table 2 are obtained. The values of the osmotic coefficient listed under the heading ϕ_{obs} were interpolated from the measured values of Rush and Johnson.¹⁰ As can be seen, the agreement between the values predicted using equation (3), listed under the heading ϕ_c , and the observed values, is very good.

¹⁰R. M. Rush and J. S. Johnson, J. Chem. Eng. Data 11, 590 (1966).

Table 1. Values of the parameters of equations (2) and (2a)
as estimated by least squares

	HCl	NaCl	MgCl ₂	MgSO ₄
p ₁	- 0.878725	6.78376	- 0.178750	- 14.5582
p ₂	83.5061	- 357.476	64.9074	795.248
p ₃	0.129604	- 0.975765	0.00881813	2.07668
p ₄	5.83637	- 0.657028	0.317576	2.94534
p ₅	- 286.750	38.5245	- 17.4918	- 156.753
p ₆	- 0.855136	0.093631	- 0.0447522	- 0.423531

Two separate determinations of the osmotic coefficient of synthetic sea water have been reported at elevated temperatures. Gibbard and Scatchard⁴, using a synthetic sea water of essentially the same composition as that used by Rush and Johnson¹⁰, measured values of the osmotic coefficient from 25-100°; while Liu and Lindsay⁵ reported values from 75-300°. Both the Rush and Johnson and the Gibbard and Scatchard formulations for synthetic sea water contained K⁺ ion with the m_{K^+}/m_{Na^+} ratio of about .021. Since the osmotic coefficient of KCl solutions has not been measured at elevated temperatures the K⁺ was replaced with Na⁺ in our calculations. This substitution was also made in the calculations at 25°. The excellent agreement between the observed and calculated osmotic coefficients displayed in Table 2 demonstrates that this substitution is at most a very small source of error in the calculated values.

If the K⁺ is replaced by Na⁺ in the Gibbard-Scatchard formulation for synthetic sea water the NaCl, MgCl₂, and MgSO₄ appear in the ratio 1 : 0.0698 : 0.0589. Values of the osmotic coefficient of a synthetic sea water of

Table 2. Osmotic coefficients of artificial sea water at 25°
 NaCl-MgCl₂-MgSO₄ in ratio 1 : 0.0745 : 0.0586

I	ϕ_{obs}	ϕ_{c}^*
0.5	0.9018	0.8993
0.7	0.9062	0.9031
1.0	0.9155	0.9128
2.0	0.9619	0.9616
3.0	1.0232	1.0250
4.0	1.0938	1.0979
5.0	1.1716	1.1771
6.0	1.2536	1.2597

*The small amount of K⁺ present in the solutions used to measure ϕ_{obs} was replaced by Na⁺ in computing ϕ_{c} (see page 5).

this composition at 75 and 100°, calculated using the parameters in Table 1 along with the equations (1) and (3), are shown in Table 3. Again the agreement between the calculated values and the measured values of Gibbard and Scatchard is very good at all three values of the total ionic strength, with the greatest deviation between the measured and observed values being about one percent at 100° at an ionic strength of 5.807.

Similar calculations for the synthetic sea water used by Liu and Lindsay with NaCl-MgCl₂-MgSO₄ in the ratio 1 : 0.0773 : 0.0613 at temperatures from 75 to 300° yield the osmotic coefficient values shown in Table 4. Note that in this case the observed values are considerably lower than the calculated values at the lower values of the ionic strength and temperature. Since the agreement between the calculated and observed values is so good

Table 3. Osmotic coefficients of artificial sea water at 75 and 100°
 $\text{NaCl-MgCl}_2\text{-MgSO}_4$ in ratio 1 : 0.0698 : 0.0589

$t^\circ\text{C}$	I	ϕ_{obs}^*	ϕ_c
75	1.014	0.9111	0.917
	2.802	1.0021	1.011
	5.795	1.2029	1.193
100	1.016	0.9038	0.903
	2.807	0.9913	0.994
	5.807	1.1708	1.159

* Ref. 4

in the case of the Gibbard-Scatchard experimental values, we suggest that the latter values of the osmotic coefficients are to be preferred for synthetic sea water over the Liu and Lindsay values at the lower temperatures.

THE ACTIVITY COEFFICIENT OF EACH COMPONENT IN HCl-NaCl-MgCl_2 MIXTURES FROM 25-55°

The activity coefficient of HCl in aqueous HCl-NaCl-MgCl_2 mixtures¹¹ has been measured by the emf technique in the temperature range 25-70°. By combining the emf measurements on the above system with isopiestic and glass electrode measurements on the system NaCl-MgCl_2 ¹² it is possible to calculate also the activity coefficients of the NaCl and the MgCl_2 in the

¹¹ M. H. Lietzke and R. J. Herdclotz, J. Inorg. Nucl. Chem. 33, 1649 (1971).

¹² J. N. Butler and R. Huston, J. Phys. Chem. 71, 4479 (1967); R. F. Platford, ibid. 72, 4053 (1968); R. D. Lanier, ibid. 69, 3992 (1965).

Table 4. Osmotic coefficients of artificial sea water from 75 to 300°
 $\text{NaCl-MgCl}_2\text{-MgSO}_4$ in ratio 1 : 0.0773 : 0.0613

$t^\circ\text{C}$	I	ϕ_{obs}^*	ϕ_c
75	2.0437	0.9160	0.9585
	4.1739	1.0653	1.0860
100	1.0682	0.8222	0.9042
	2.0474	0.9218	0.9499
	4.1897	1.0554	1.0683
125	1.0699	0.8385	0.8919
	2.0547	0.9156	0.9342
	4.2208	1.0449	1.0415
150	1.0729	0.8366	0.8753
	2.0679	0.8977	0.9123
	4.2772	1.0136	1.0079
175	1.0778	0.8240	0.8548
	2.0905	0.8742	0.8855
	4.3782	0.9760	0.9690
200	1.0856	0.8097	0.8305
	2.1265	0.8457	0.8537
	4.5415	0.9325	0.9252
225	1.0974	0.7810	0.8017
	2.1829	0.8119	0.8166
	4.8071	0.8864	0.8768
250	2.2713	0.7660	0.7728
	5.2447	0.8327	0.8236

Table 4. (Continued)

$t^{\circ}\text{C}$	I	ϕ_{obs}^*	ϕ_c
275	2.4130	0.7125	0.7195
	5.9914	0.7700	0.7666
300	2.6524	0.6515	0.6509
	7.3224	0.7052	0.7135

* Ref. 5

mixtures. Since the latter measurements have been carried out only at 25° , values of the activity coefficients of the NaCl and the MgCl_2 in the mixtures have previously been reported at this temperature only. The equations used to calculate the activity coefficients of the HCl, NaCl, and MgCl_2 in the HCl-NaCl- MgCl_2 mixtures are given elsewhere¹¹ and will not be repeated here. Since the values of the parameters in these equations were estimated by least squares by fitting activity or osmotic coefficients derived from experimental emf or isopiestic data, we may consider the activity coefficients so derived as "fitted observed values". Caution must be exercised, however, in accepting such values as "true" values, since, as pointed out in another communication¹³, the values themselves depend somewhat on the model used to derive the equations expressing the functional dependence of the activity coefficients on temperature, total ionic strength, and fraction of each component in the mixtures. Thus the value of the logarithm of the activity coefficient of HCl, for example, in a particular HCl-NaCl- MgCl_2 mixture may vary by more than a percent depending upon what model was used in fitting the experimental values. The same thing is true of the values calculated for the activity coefficients of the NaCl and the MgCl_2 in the HCl-NaCl- MgCl_2 mixtures.

¹³M. H. Lietzke and R. W. Stoughton, J. Tenn. Acad. Sci., in press.

In order to obtain "fitted observed" values of the activity coefficients of NaCl and MgCl_2 in HCl-NaCl- MgCl_2 mixtures above 25° the following procedure was used. Since NaCl- MgCl_2 solutions have not been investigated experimentally above 25° osmotic coefficients were predicted at a number of temperatures and total ionic strengths, with varying ratios of NaCl/ MgCl_2 using the parameters in Table 1 and equations (1) and (3). These osmotic coefficient values were then fitted by least squares using equation (4).

$$\phi_m' = 1 + \frac{I}{\sum v_i m_i} \left\{ \frac{-2.34404}{(1.5)^3 I} \left[1 + 1.5\sqrt{I} - 2 \ln (1 + 1.5\sqrt{I}) - \frac{1}{1+1.5\sqrt{I}} \right] \right. \\ \left. + (a_2^{(1)} + a_2^{(2)} I) I y_2 + (a_3^{(1)} + a_3^{(2)} I) I y_3 + (b_{23}^{(0,1)} + b_{23}^{(0,2)} I) I y_3 (1 - y_3) \right\} \quad (4)$$

In this equation ϕ_m' represents the osmotic coefficient of a solution containing two electrolytes indexed 2 and 3; v_i is the number of ions into which the i th electrolyte at molality m_i in the mixture dissociates; y_i is the ionic strength fraction (I_i/I) of the i th electrolyte in the mixture; and I is the total ionic strength of the solution. The parameters $a_2^{(1)}$, $a_2^{(2)}$, $a_3^{(1)}$, and $a_3^{(2)}$ are respectively the parameters giving the dependences of the osmotic coefficients of electrolytes 2 and 3 (subscripts) on the first and second power of the ionic strength (superscripts), while $b_{23}^{(0,1)}$ and $b_{23}^{(0,2)}$ represent interaction coefficients between the two electrolytes.¹⁴ While ϕ_m' represents the same quantity as does ϕ_m in equation (3), they will not in general be equal for they are based on different theoretical models for mixtures.

Values of the parameters in equation (4) obtained by fitting the predicted values of the osmotic coefficient of the NaCl- MgCl_2 mixtures were combined with the parameters obtained by fitting the $\log \gamma_{\text{HCl}}$ values calculated from the experimental emf values to calculate "fitted observed" values of $\log \gamma_{\text{NaCl}}$ and $\log \gamma_{\text{MgCl}_2}$ in the HCl-NaCl- MgCl_2 mixtures. The "fitted observed" values of $(\log \gamma + 1)$ for HCl, NaCl, and MgCl_2 in mixtures with values of the total ionic strength one and two at temperatures 35° , 45° , and 55° are shown in columns six, eight, and ten of Table 5. Values of the same quantities predicted solely from the osmotic coefficients of the

¹⁴G. Scatchard, J. Am. Chem. Soc. 83, 2636 (1961).

Table 5. Activity coefficient values in HCl-NaCl-MgCl₂ mixtures
at temperatures above 25°

t°C	I	X ₂	X ₃	X ₄	log γ + 1					
					HCl _(obs)	HCl _(c)	NaCl _(obs)	NaCl _(c)	MgCl ₂ (obs)	MgCl ₂ (c)
35	1	0.750	0.125	0.125	0.907	0.903	0.876	0.858	0.712	0.722
	1	.500	.250	.250	.898	.892	.859	.847	.692	.701
	1	.250	.375	.375	.889	.881	.842	.836	.674	.679
	2	.750	.125	.125	.995	.992	.935	.901	.797	.813
	2	.500	.250	.250	.979	.970	.902	.879	.754	.769
	2	.250	.375	.375	.961	.948	.868	.857	.715	.725
45	1	.750	.125	.125	.903	.897	.874	.855	.703	.709
	1	.500	.250	.250	.894	.886	.857	.844	.683	.689
	1	.250	.375	.375	.885	.876	.840	.834	.664	.668
	2	.750	.125	.125	.988	.985	.932	.899	.782	.797
	2	.500	.250	.250	.973	.964	.898	.878	.740	.753
	2	.250	.375	.375	.955	.942	.866	.856	.702	.710
55	1	.750	.125	.125	.898	.890	.869	.851	.703	.696
	1	.500	.250	.250	.890	.880	.852	.841	.680	.676
	1	.250	.375	.375	.882	.869	.836	.831	.657	.656
	2	.750	.125	.125	.979	.976	.924	.895	.783	.778
	2	.500	.250	.250	.966	.955	.891	.874	.736	.736
	2	.250	.375	.375	.950	.934	.860	.853	.692	.694

individual components of the mixtures are shown in columns seven, nine, and eleven of Table 5. As can be seen the agreement between the two sets of values is remarkably good. Similar calculations have been made on other multicomponent electrolyte systems with similar results. Hence, it appears that if good osmotic coefficient data are available on the individual components of an aqueous electrolyte mixture it is possible to predict to within a percent or two the activity coefficients of each component in the mixture.

CONCLUSIONS

The examples presented in this report demonstrate that simple equations developed for predicting osmotic and activity coefficients for aqueous electrolyte mixtures at 25° also can be used for predictions at elevated temperatures. If the osmotic coefficient of a mixed electrolyte solution is needed, then equation (3) may be used, providing, of course, that the osmotic coefficient of each component in the mixture is known at the desired temperature. If the activity coefficient of each component in a mixed electrolyte solution is required, then two different approaches may be used. In the simplest, one takes advantage of the fact that the parameters of the equation used to fit the osmotic coefficients of solutions of each individual component are directly related to the parameters of the equation for the activity coefficient of the solute.¹⁵ Hence, activity coefficient values are computed for each component of the mixture from the osmotic coefficients and these activity coefficient values then combined appropriately using one of the methods cited in reference (2). Alternatively, one could predict a set of osmotic coefficients for a given mixture using equation (3) by varying the total ionic strength of the solution and the fractions of each component. The predicted osmotic coefficients can then be fitted by least squares using equation (4) to obtain estimates of the parameters of the equation. The activity coefficient of each component in the mixture may then be computed for a given set of conditions directly from the

¹⁵M. H. Lietzke and R. W. Stoughton, J. Phys. Chem. 66, 508 (1962).

parameters of equation (4). As the calculations on the HCl-NaCl-MgCl₂ mixtures illustrate these two methods give very similar results.

As mentioned previously, only the osmotic coefficients of HCl, NaCl, MgCl₂, and MgSO₄ have been measured accurately and carefully to elevated temperatures. If predictions are to be made of osmotic or activity coefficients for geothermal solutions, then the osmotic coefficient of a few additional electrolyte solutions will need to be measured at elevated temperatures. The most important of these are KCl and CaCl₂. With these additional data it should be possible to predict closely both osmotic and activity coefficients for geothermal brines at temperature.

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